

MOLECULAR THEORY OF NON-NEWTONIAN FLUIDS

Status Report for September 16, 1965 to March 15, 1966

Grant Number: NsG-590/48-02-016

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Objective: The objective of this research program is to develop theoretical methods for predicting and correlating the rheological behavior of non-Newtonian suspensions and polymer solutions. Our investigation, which is similar in many respects to the molecular theory of Newtonian fluids, is based on determining the fundamental mechanisms governing momentum transport in suspensions and polymer solutions in order to relate the viscosity coefficients of these fluids to the characteristic parameters of their microstructure. Given values of these parameters, which include the size, shape, flexibility, mass distribution and concentration of the components of the fluid, one can calculate flow rates, velocity profiles, pressure drops and other desired information about the flow.

Background: The essential structural feature of all non-Newtonian fluids which distinguishes them from Newtonian fluids is the presence of large molecules or particles. These drastically modify the flow from what it would be were they not there by inducing long-range spatial correlations in the fluid velocity field. The size, shape, flexibility, and concentration of the particles determine the nature and extent of the correlation; e.g., the correlation is more extensive and has a shorter relaxation time for rigid particles than for flexible ones. Thus, in order to develop a theory adequate to account for the variety of rheological behavior observed with particulate fluids, it is necessary to consider the dynamics of large molecules in solution or suspension and to relate the dynamical behavior to the overall flow characteristics of the material.

Many investigations of the structural basis of non-Newtonian flow have been undertaken, but they are of limited usefulness because of restrictive assumptions made in the analyses, principally that the concentration of particles is small enough that interactions can be neglected. The theories

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so obtained bear a marked similarity to the kinetic theory of dilute gases. This is instructive because it points out the need for a molecular theory of particulate fluids analogous to the kinetic theory of solids, that is, a molecular theory of highly concentrated suspensions and polymer solutions. One could then blend the essential features of both the dilute and concentrated cases, in much the same manner as is done in the kinetic theory of dense gases and liquids, to obtain a workable theory for the whole range of concentrations. Any analysis incorporating the two viewpoints is bound to be more potent than one based on either one or the other. To this end we are attempting to develop a kinetic theory of highly concentrated particulate fluids.

Progress and Results: We are continuing our investigation of the rheology of concentrated suspensions of rigid particles within the framework of the layer model described in the thesis referenced below. This model is based on the fact that the rate of momentum transport by interparticle collisions amounts to a small fraction of actual measured rates. Therefore, it is our hypothesis that the predominate effect of collisions is to establish more or less well-defined layers of particles within a flowing suspension. The presence of these layers presumably increases the actual local velocity gradient in the fluid layer between the particle layers above the measured local velocity gradient, which is an average over a region large compared with the layer dimension. This results in a viscosity which is larger than that of the suspending fluid. It is our belief that the thickness of the fluid layers increases when the overall shear rate is increased, approaching a limiting value asymptotically as the shear rate becomes larger and larger. This behavior is reflected in a suspension viscosity which decreases monotonically with shear rate to an asymptotic value in accord with the observed behavior of suspensions of rigid particles. We have been examining the factors which determine the layer spacing of such a model, and have found qualitatively that the spacing does increase with shear rate. We are presently carrying out a detailed analysis of the layer mechanism which utilizes a variational principle based on the concept of minimum rate of energy dissipation. This will allow us to ascertain quantitatively how well the model predicts the variation of suspension viscosity with shear rate and particle concentration.

Plans: After completing the work on the utility of the variational method mentioned above, we plan to investigate the compatability of the results of our model with the "power law" form of the momentum constitutive relation and with the general Stokesian form obtained from continuum mechanics. Next we plan to attempt a detailed statistical mechanical treatment of the particle motion in flowing suspensions. This will require the derivation and solution of the appropriate form of the Boltzmann equation, which determines the distribution function specifying the probability of particular velocity states. The Boltzmann equation will necessarily contain the velocity-dependent drag force experienced by a particle as it moves through the viscous suspending medium. Beyond this we plan to examine the physical, structural basis of the rheology of concentrated flexible-particle suspensions and polymer solutions in the light of our work on suspensions of rigid particles.

Reference: "The Rheology of Concentrated Suspensions", by Robert K. Ham, M.S. Thesis, University of Washington (1965).

Date: March 25, 1966